Appl. Phys. B 74, 307–311 (2002)
DOI: 10.1007/s003400200811

Applied Physics B
Lasers and Optics

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Two-octave spectral broadening of subnanojoule Cr:forsterite femtosecond laser pulses in tapered fibers

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Received: 23 October 2001 / Accepted: 16 January 2002
Published online: 14 March 2002 •

ABSTRACT Spectral broadening of femtosecond Cr:forsterite laser pulses is enhanced due to the use of tapered fibers. Supercontinuum generation with unamplified subnanojoule femtosecond Cr:forsterite laser pulses is observed for the first time. With 40-fs 0.6-nJ pulses of 1.25-μm Cr:forsterite laser radiation coupled into a tapered fiber having a taper waist diameter of about 2 μm and a taper waist length of 90 mm, we observed the spectra spanning more than two octaves at the output of the fiber in the regime of anomalous group-velocity dispersion. This result opens the way for the creation of compact femtosecond Cr:forsterite laser plus tapered fiber systems for optical metrology and biomedical applications.

PACS 42.65.Wi; 42.81.Qb

1 Introduction

Spectral-broadening and supercontinuum-generating abilities of optical fibers have been recently greatly enhanced through fiber microstructuring and tapering. The use of holey [1–7] and then tapered [8] fibers allowed supercontinuum to be generated [8, 9] starting with very low (typically subnanojoule) levels of energies of femtosecond laser pulses. This has already resulted in a major breakthrough in optical metrology [10–13], based on the ideas of using frequency combs produced by mode-locked lasers for high-precision measurements [14], allowing the creation of compact frequency chains phase-coherently linking radio-frequency reference sources to the optical region through frequency combs generated by femtosecond mode-locked lasers, which are then spectrally broadened to span more than an octave in a holey [10–14] or a tapered [15] fiber. Supercontinuum generation in holey and tapered fibers also offers the way to create new broadband sources for spectroscopic applications and suggests a new approach to pulse compression [8, 9]. The application of such fibers in telecommunication technologies [3, 4] is another very important issue, which is being actively explored at the moment.

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Physically, the capability of holey and tapered fibers to enhance nonlinear optical processes is due to the small radius of the fiber core, permitting high intensities of laser radiation to be achieved [3–6, 8]. These fibers also provide robust single-mode waveguiding within a broad spectral range [2] and allow very high contrasts of refractive indices in the core and the cladding to be attained, thus resulting in a high degree of light localization in the fiber core [2, 6, 8, 16]. Finally, the tunability of dispersion in holey [9] and tapered [8] fibers makes them an extremely useful tool for ultrafast optics, including the transmission, frequency conversion (e.g. through efficient harmonic generation [17]), compression, and spectral control of ultrashort laser pulses, even opening the way for subfemtosecond fiber optics.

While most of the nonlinear optical experiments with holey and tapered fibers, including supercontinuum generation [8, 9] and high-precision measurements with femtosecond frequency combs [10–15], have been performed with Ti:sapphire laser pulses, it is of considerable interest to adapt this new fiber-optic technique to femtosecond lasers generating radiation with longer wavelengths. In particular, the use of holey and tapered fibers to spectrally broaden and frequency-convert femtosecond Cr:forsterite laser pulses is of special interest, since these lasers are as compact and convenient as Ti:sapphire systems, covering, at the same time, the wavelength range that is very promising for biomedical applications and that opens the way, in principle, to reduce the number of frequency-division stages in establishing a link with methane-stabilized 3.39-μm He–Ne lasers in femtosecond optical metrological systems.

In this paper, we present the results of our experiments devoted to the investigation of the spectral broadening of femtosecond Cr:forsterite laser pulses in tapered fibers. These studies allowed us to enhance the spectral broadening of unamplified subnanojoule femtosecond Cr:forsterite laser pulses and to observe for the first time supercontinuum generation with such pulses, giving rise to spectra spanning more than two octaves.

2 Experimental

Our laser system (Fig. 1) was based on a Nd:YAG-pumped Cr^{4+}:forsterite oscillator [18], which used a 19-mm

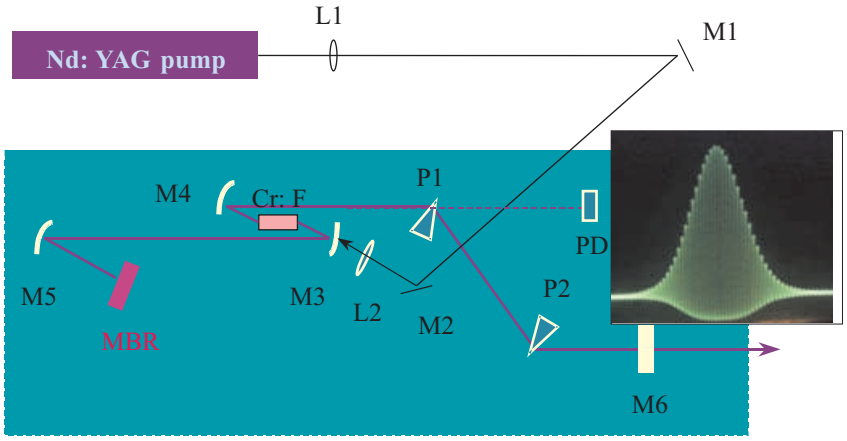

FIGURE 1 Diagram of the all-solid-state femtosecond Cr^{4+}:forsterite laser: Nd:YAG pump, Spectra Physics Millennia Nd:YAG laser; MP1, MP2, pump mirrors; L1, L2, lenses; M3–M5, cavity mirrors; P1, P2, intracavity prisms; Cr : F, $Cr^{4+} : Mg_2SiO_4$ crystal; M6, output coupler; MBR, multilayer Bragg reflector; PD, photodetector. The inset shows an autocorrelation trace of a Cr:forsterite laser pulse with a duration of approximately 30 fs

Cr^{4+}:forsterite crystal, focusing mirrors with a radius of curvature equal to 100 mm, and a 4.5% output coupler. As an option, a semiconductor saturable-absorber reflector could be used as a rear cavity mirror. Self-starting mode locking in this system was achieved both with and without semiconductor saturable-absorber mirrors. The laser was capable of generating pulses with a duration of 30 to 100 fs. A typical autocorrelation trace of a 30-fs output pulse of the Cr:forsterite laser is shown in the inset in Fig. 1. With a 6–9-W pump from a Spectra Physics Millennia Nd:YAG laser, our Cr:forsterite laser generated 450 mW of average output power. Frequency doubling in the developed laser systems can be implemented, as described in [19], through second-harmonic generation in a 2.5-mm-long deuterated cesium dihydrogen arsenate (DCDA) crystal, ensuring group-velocity matching for femtosecond pulses produced by the Cr:forsterite laser.

Unamplified radiation produced by the Cr:forsterite laser was coupled into a tapered fiber (Fig. 2), manufactured by tapering Corning SMF-28 standard telecommunications fibers with a core diameter of $\sim 9\,\mu$m, a cutoff wavelength of 1250 nm, and a numerical aperture of 0.1. The fiber-tapering procedure was described in detail in [8]. This process involved fiber tapering by means of heating and stretching in a flame. A narrow taper waist was connected to untapered fibers by taper transitions. The residual fiber core in the narrow taper waist is negligibly small, so the light fills the whole fiber in the waist. However, the large refractive-index step between silica and air allows the light to be strongly confined within the fiber even if the diameter is as small as 2 μm, increasing the intensity and, thus, enhancing the efficiency of nonlinear optical processes in such fibers. In particular, the enhancement of self-phase modulations in such fibers has been earlier demonstrated by Stegeman's group [20].

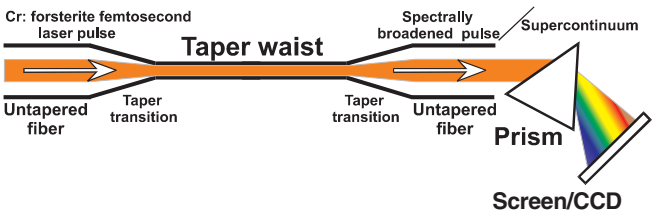

FIGURE 2 Diagram of Cr:forsterite pulse propagation through a tapered fiber. The prism is used to spectrally disperse the output beam

3 Results and discussion

The spectra of 70-fs pulses of different energies coming out of a tapered fiber with $\sim$ 35-mm transitions and a 90-mm waist of a uniform diameter of $\sim 2\,\mu$m are shown in Fig. 3. Tapered fibers with such a waist diameter provide an anomalous group-velocity dispersion for the fundamental radiation of the Cr:forsterite laser (Fig. 4), with a zero group velocity dispersion being achieved around 700 nm. This situation is favorable for the nonlinear propagation of ultrashort Cr:forsterite laser pulses and for supercontinuum generation in the visible range. We were able to achieve a considerable spectral broadening even with 100-pJ light pulses coupled into the fiber. On increasing the energy of the laser pulses, we observed the growth in the bandwidth of pulses coming out of the fiber (Fig. 3). To get an order-of-magnitude estimate of the characteristic parameters of the process, we approximate the dependence of the spectral broadening $\Delta\omega$ on the energy of

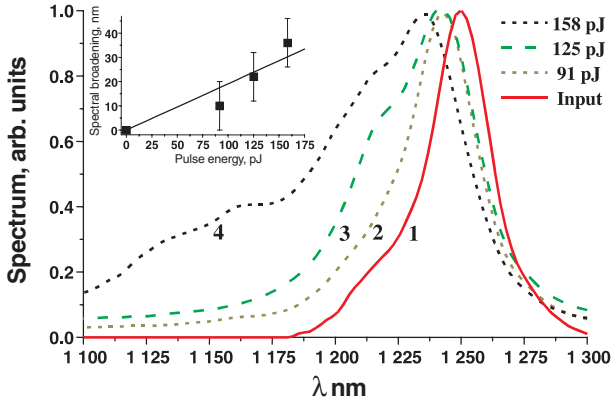

FIGURE 3 Spectral broadening of subnanojoule 70-fs pulses in a tapered fiber with $\sim$ 35-mm transitions and a 90-mm waist of a uniform diameter of $\sim 2\,\mu$m. *Solid line* 1 show the spectrum of the input pulse. *Lines* 2–4 display the spectra of the pulses at the output of the fiber with (2) 0.09, (3) 0.13, and (4) 0.16 nJ of pulse energy coupled into the fiber. The inset shows the spectral broadening as a function of the pulse energy coupled into the fiber approximated with a linear dependence

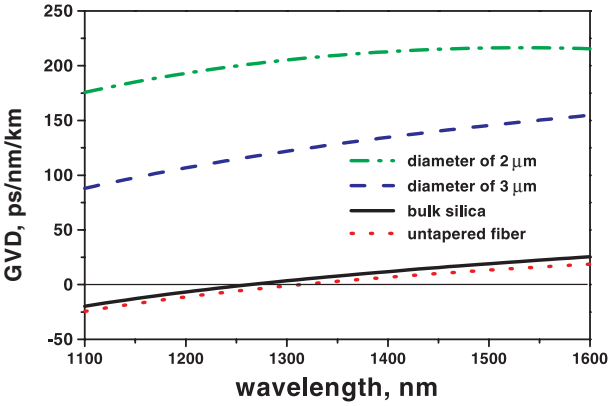

FIGURE 4 Group-velocity dispersion calculated for a (*dash-dotted line*) 2-μm and a (*dashed line*) 3-μm fused silica fiber taper waist with an air cladding, (*solid line*) bulk silica, and (*dotted line*) a standard untapered fiber with a core diameter of 9 μm and 0.36% core–cladding refractive-index step

laser pulses coupled into the fiber with a linear function (see the inset in Fig. 3). This idea is based on the elementary theory of self-phase modulation, which gives the following expression for the relative spectral broadening [21]: $\Delta\omega \approx \gamma PL/\tau$, where γ is the nonlinear coefficient, P is the pulse power, L is the length of the working area of the fiber, and τ is the pulse duration. In this limit, the nonlinear coefficient γ of our fiber can be estimated as $20\ \mathrm{W}^{-1}\ \mathrm{km}^{-1}$, indicating a very high efficiency of nonlinear optical processes in the fiber. Although very crude, this estimate of the nonlinear coefficient γ is very useful, as it gives us an impression of the spatial scale of nonlinear optical processes in a tapered fiber, thus allowing the influence of various nonlinear optical effects on spectral broadening and spectral distortions of short laser pulses in the fiber to be assessed.

Although the initial stage of spectral broadening considered above does not lead to very large spectral widths, it is very useful for many applications due to the fact that the spectral width of femtosecond pulses can be easily and reproducibly controlled by changing the energy of input pulses. Femtosecond Cr^{4+}:forsterite lasers are currently intensely discussed as attractive sources for a broad range of medical applications [18, 22–25]. Due to the longer wavelength, the scattering cross section of Cr^{4+}:forsterite laser radiation in biological tissues is much less than that typical of Ti:sapphire lasers, while absorption of biological tissues is reasonably low within the wavelength range covered by Cr^{4+}:forsterite lasers. All-solid-state Cr:forsterite lasers have recently been shown to be an optimal choice for high signal-to-noise ratio optical coherence tomography (OCT) imaging, allowing a resolution of several microns to be achieved [22, 23]. In this context, the above-described regime of spectral broadening seems to be very useful for the creation of compact Cr:forsterite laser systems for optical coherence tomography, where the coherence length and, hence, the spatial resolution can be tuned through an extracavity chirp control with the use of tapered fibers.

The spectral broadening of Cr:forsterite laser pulses in a tapered fiber was accompanied by pulse chirping. This can be clearly seen from second-order autocorrelation traces measured for different input pulse energies (Fig. 5) using the two-photon absorption process on the silicon surface of a photodetector. Autocorrelation traces become noticeably

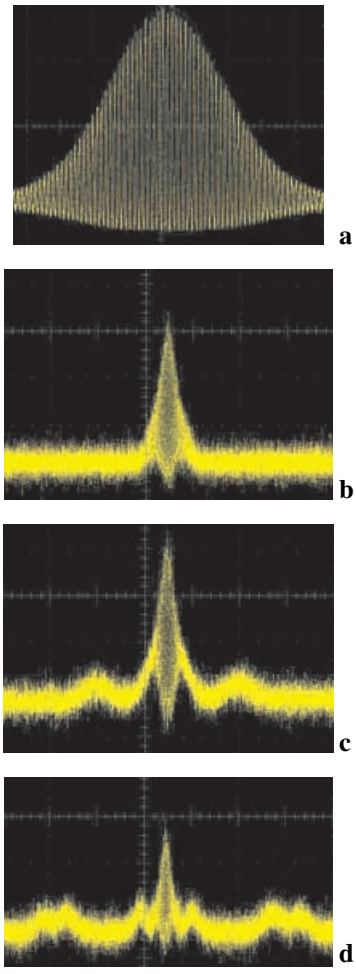

FIGURE 5 Second-order autocorrelation traces of **a** an 80-fs Cr:forsterite pulse at the input of the fiber and pulses coming out of the fiber with a pulse energy of **b** 40 pJ, **c** 100 pJ, and **d** 130 pJ. Pulse energies are measured at the output of the fiber

distorted with the increase in the energy of Cr:forsterite pulses coupled into the fiber. Intensity-dependent features in these traces indicate pulse chirping due to nonlinear optical processes in the fiber (cf. Fig. 5a–d). Although the length of the untapered fiber sections was much larger than the length of the taper waist, nonlinear optical effects were easily detectable even with sub-0.1-nJ 80-fs laser pulses (Fig. 5b), which become possible due to the enhanced nonlinear optical interactions in the fiber waist and a very low group-velocity dispersion of the untapered sections of the fiber at the wavelength of Cr:forsterite laser radiation (the dotted line in Fig. 4). Although these autocorrelation measurements, of course, do not allow the phase information to be extracted in an unambiguous way, the results of these experiments demonstrate that the employed combination of the tapered-fiber component with femtosecond Cr:forsterite laser pulses is advantageous for a full, phase-sensitive characterization of the process of spectral broadening of femtosecond pulses in a tapered fiber. This is because the linear phase shift related to the group-velocity dispersion of the untapered sections of the fiber is small in our arrangement, while the nonlinear, intensity-dependent phase shift can be easily observed with very low energies of laser

pulses. A full characterization of the output pulses, including the extraction of the spectral phase distribution, would be of considerable importance for understanding the ways of using tapered fibers for pulse compression and then for optimizing tapered-fiber pulse compressors. Frequency-resolved optical gating [26] seems to be an adequate technique for such investigations, in view of the favorable relation between the linear and nonlinear phase shifts, demonstrated by our experiments. Such frequency-resolved studies, however, fall beyond the scope of this paper.

Starting with a pulse energy of 0.6 nJ, a broad continuum emission was observed in our experiments when 40-fs pulses of 1.25-μm Cr:forsterite laser radiation were coupled into a tapered fiber with the above-specified parameters. The Cr:forsterite laser generated 40-fs pulses of an average power of 250 mW at a repetition rate of 120 MHz in these experiments, with approximately 70–75 mW of average power being coupled into the tapered fiber. A detailed characterization of the broad spectrum of radiation coming out of the tapered fiber under these conditions falls beyond the scope of this paper, as the optimal regime of supercontinuum generation, providing the maximum flatness of this radiation, still has to be found. However, the width of the spectrum generated in our experiments can be estimated from Fig. 6, which shows a digital-camera image of the output light beam dispersed with an SF-6 prism and visualized on a white screen, helping to appreciate the wealth of new colors in the spectrum of a subnanojoule light pulse coming out of the fiber. The upper image is taken with the camera located at a distance of 40 cm from the screen. The image is, therefore, well focused, but the sensitivity of the camera is not enough to reproduce the red light (around 700 nm, the area 1 in Fig. 6). The lower image is taken with the camera located at a distance of 10 cm from the screen. The red part of the spectrum is clearly seen in this case, but the image is unfocused. The spectrum shown in Fig. 6 spans more than two octaves, stretching beyond the spectral area around 417 nm (region 2 in Fig. 6), characteristic of the third harmonic of input radiation (easily identifiable with the third harmonic of our Cr:forsterite laser produced with a nonlinear crystal), and giving rise to intense luminescence of the screen in the area of wavelengths less than roughly 400 nm, where luminescence is never observed for longer wavelengths (region 3 in Fig. 6). No supercontinuum generation was observed when subnanojoule Cr:forsterite laser pulses were propagated through untapered fibers.

It is very instructive also to study in greater detail the intermediate stage between the regime of small spectral broadening (Fig. 3) and the regime of supercontinuum generation (Fig. 6). This stage reveals that supercontinuum generation

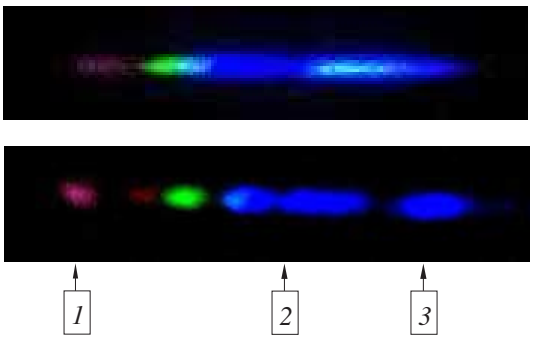

FIGURE 6 Superbroadening of a 40-fs 0.6-nJ pulse of 1.25-μm Cr:forsterite laser radiation in a tapered fiber. The output light beam is dispersed with an SF-6 prism and visualized on a white screen: (1) visible red (around 700 nm), (2) the third-harmonic area around 417 nm, and (3) the area of screen luminescence corresponding to wavelengths less than 400 nm. A digital camera is located at a distance of 40 cm (the upper image) and 10 cm (the lower image) from the screen

under our conditions is a result of a rather complicated interplay of different nonlinear optical processes. In particular, spectral distortions and asymmetry noticeable in spectrum 4 in Fig. 3, which corresponds to 70-fs pulses of a Cr:forsterite laser with a pulse energy of approximately 0.16 nJ, may be indicative of the steepening of the trailing edge of the pulse due to the intensity dependence of the group velocity in a nonlinear medium, which may result in shock-wave formation [21] in the envelope of a light pulse at higher laser intensities.

In a simple approximation when dispersion effects are negligible, the evolution of the intensity I of an initially Gaussian pulse with an intensity-dependent group velocity is governed by [21] $I(\xi,\theta)=\exp\left[-(\theta-3I\xi T_0/\pi\tau)^2\right]$, where $\xi=z/L_{\mathrm{nl}}$, z is the coordinate along the fiber, $L_{\mathrm{nl}}=1/\gamma P$ is the characteristic length of nonlinear interaction, θ is the running time, T_0 is the duration of the optical cycle, and τ is the pulse duration. Using the estimate for the nonlinear coefficient γ obtained above, we find that the critical length of shock-wave formation (i.e. the length where the derivative of the pulse envelope in the running time becomes infinity) for $P\approx 2.3$ kW can be estimated as [21] $L_{\mathrm{sh}}\simeq 1.2\tau/\gamma P T_0\approx 30$ cm (T_0 is the duration of the field cycle). As a result, spectral distortions due to the steepening of the trailing edge of the pulse may become noticeable for fiber samples employed in our experiments, and the shock-wave formation may be one of the processes involved in supercontinuum generation. This is consistent with earlier observations by Ranka et al. [9] and Fedotov et al. [27].

Table 1 puts the results of our experiments in the context of other works on supercontinuum generation in microstruc-

	Holey fibers				Tapered fibers			
	τ (fs)	W (nJ)	L (cm)	Ref.	τ (fs)	W (nJ)	L (cm)	Ref.
Ti:sapphire laser	100	0.8	75	[9]	200	3.9	9	[8]
Cr:forsterite laser	Spectral broadening of 30-fs unamplified pulses was observed in [28]				40	0.6	9	This work

Notation: τ is the pulse duration, W is the pulse energy, and L is the propagation length

TABLE 1 Supercontinuum generation with unamplified femtosecond laser pulses

ture and tapered fibers. The possibility of supercontinuum generation by propagating subnanojoule-level Ti:sapphire laser pulses through microstructure fibers, demonstrated by Ranka et al. [9], has opened new avenues in many areas of ultrafast optics and spectroscopy, allowing important problems of optical frequency metrology to be solved [10–14]. The search for a simpler design of a fiber capable of generating a supercontinuum with low-power laser pulses has brought Birks et al. [8] to an idea of using tapered fibers for this purpose. This concept was successfully demonstrated by supercontinuum generation with unamplified Ti:sapphire laser pulses reported in [8]. Self-phase modulation of unamplified 30-fs Cr:forsterite laser pulses and the generation of cross-phase-modulated third harmonics with the use of such pulses have been recently observed in holey fibers [28]. In the present paper, we communicated for the first time supercontinuum generation with unamplified femtosecond Cr:forsterite laser pulses propagating through a tapered fiber and demonstrated that subnanojoule energies of femtosecond Cr:forsterite laser pulses may be sufficient, with an appropriate choice of pulse durations and fiber dispersion, to generate light with a spectrum spanning more than two octaves.

4 Conclusions

Thus, we detected supercontinuum generation with unamplified subnanojoule femtosecond Cr:forsterite laser pulses. Coupling 40-fs 0.6-nJ pulses of 1.25-μm Cr:forsterite laser radiation into a tapered fiber with a taper waist diameter of about 2 μm and a taper waist length of 90 mm, we ended up with the spectra spanning more than two octaves at the output of the fiber in the regime of anomalous group-velocity dispersion. Our measurements have also demonstrated that supercontinuum generation in tapered fibers may be accompanied by a rather complicated combination of nonlinear optical processes which, in principle, may lead to the degradation of coherence in the supercontinuum, thus lowering the usefulness of this approach in optical frequency metrology, as well as in ultrafast and biomedical optics. A careful analysis of these factors is, therefore, necessary to assess their influence on the properties of supercontinuum and to understand the requirements of the fiber dispersion and laser-pulse characteristics to reduce the influence of these effects or to precompensate for them. Enhanced spectral broadening and supercontinuum generation from unamplified pulses of a Cr:forsterite laser observed in our experiments allow a femtosecond Cr:forsterite laser plus tapered fiber system to be proposed as a convenient and compact tool for optical metrology and biomedical applications, as well as an efficient broadband source for various spectroscopic applications.

ACKNOWLEDGEMENTS The work of DAA, ABF, AAP, AMZ, AAI, and MVA was supported in part by the President of the Russian Federation (Grant No. 00-15-99304), the CRDF (Grant Nos. RP2-2266 and RP2-2275), the Volkswagen Foundation (Project No. I/76 869), and the Russian Foundation for Basic Research (Project No. 00-02-17567). The work of DAA, ABF, AAP, AMZ, AAI, MVA, SNB, and VSP was supported through the 'Fundamental Metrology' Federal Program of the Ministry of Industry, Science, and Technology of the Russian Federation. WJW is a Royal Society University Research Fellow.

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